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REVIEW

A review on common vegetables and legumes as promising plant-based natural coagulants in water clarification

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Abstract Coagulation and flocculation provide a rather straightforward method towards water clarification. However, ongoing debates over worrying health issues linked to chemical coagulants have paved the way to develop plant-based natural coagulants. Natural coagulants are not only water clarifying agents, but they also have antimicrobial and heavy metal removal properties in some instances. These are highly attractive in the transformation of raw surface water into potable drinking water. A total of 14 plant-based natural coagulants categorized as common vegetables and legumes are identified and presented collectively in this comprehensive review. The two main coagulation mechanisms leading to the observed coagulation activities are postulated to be charge neutralization and bridging. Turbidity removal efficiencies were proven to be greatly affected by pH variations and the dosage of natural coagulants used. The existing research gaps are acknowledged in this work to provide a platform towards the necessity of further research in the water treatment processes.

Keywords Coagulation · Flocculation · Protein · Turbidity removal · Antimicrobial properties · Coagulation mechanisms

Introduction

Approximately 75 % of the universe is covered with water. Yet, some parts of the world have only limited access to this precious commodity. This irony has surfaced since the ancient times and along with mankind's progressive hunger for development; the conditions worsen especially in developing countries and rural areas. According to the statistics at UNICEF/WHO (2009), fatality cases exceeding 1.5 million are reported yearly as a result of the consumption of insanitary water leading to diarrhoea. Among which, 90 % of the cases involve children below the age of five, and in today's world, a child dies from a water-related disease every 21 s. These shocking revelations demand serious attention from all parties globally on the importance of research on water management and sanitation (Gholikandi et al. 2012; Chang et al. 2012; Chen et al. 2012; Chu et al. 2012; Lu and Huang 2012; Luo and Farrell 2013; Nagar et al. 2013; Odiyo et al. 2012; Wang and Wang 2012).

Both the surface water and groundwater provide a continuous mean of water supply to be treated into drinking water (Percival et al. 2000). These water sources could be contaminated with pathogenic microorganisms, dissolved and suspended solids, colour- and odour-causing particles rendering the water to be unsafe for direct human consumption. The presence of such impurities diminishes the quality of the water, and they must be removed effectively. Adequate water sanitation which typically comprises the coagulation and flocculation, sedimentation, filtration and disinfection processes (Ndabigengesere and Subba Narasiah 1998a) is the key to minimizing the health-threatening potentials from the related water-borne diseases.

Coagulation and flocculation

Turbid water that is murky or cloudy in appearance caused by impurities imparts an unpleasant taste to the water and thus has become the impetus behind the need for water

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treatment. Such colloidal particles are generally too fine to settle by gravity and are usually negatively charged (Kim et al. 2001; Sincero and Sincero 2002). As they are surrounded by repulsive ion charges (Fig. 1), the clumping of the individual particles is hindered. Consequently, they would form a stable suspension and could not be removed without the introduction of a coagulant. With the addition of coagulants (Step 1, Fig. 1), the repulsive ion charges would be neutralized (Step 2, Fig. 1) and particle aggregations would occur. Coagulation is typically the first step in the water treatment process. The significance of the coagulation process lies in its ability to form larger destabilized particles known as microflocs which in turn helps to remove turbidity in the water source. Apart from turbidity, pathogens and other finely dispersed colloids would also be removed to improve the water quality and subsequently leading to better human health. The chronology in the development of coagulation theory and principle has been outlined in detailed elsewhere (Jiang 2001).

Following coagulation, flocculation takes place as the adjunct to the enhancement of the microflocs formation (Bratby 2006) as indicated by steps three to five in Fig. 1. Unlike the former which occurs in <10 s, flocculation takes a longer period of approximately 20–45 min which is common in water treatment plants (Crittenden et al. 2005). Flocculation can be further classified into two stages: the first being perikinetic flocculation and the second being orthokinetic flocculation (Bratby 2006; Crittenden et al. 2005). Throughout flocculation, the size of the flocs will continue to grow until they reach the steady-state floc size distribution.

Natural coagulants

The idea of utilizing natural coagulants for the clarification of turbid water is in practice since the ancient times, even way before the advent of chemical coagulants (Ndabigengesere and Subba Narasiah 1998a; Ndabigengesere et al. 1995). Ancient civilizations in India, China and Africa are believed to have used plant derivatives as natural coagulants in their water sources since 2000 years ago (Asrafuzzaman et al. 2011). This is also evident from the Sanskrit writings in India dating back to 400 AD (Dorea 2006; Bratby 2006), the Old Testament and Roman records dating back to 77 AD (Dorea 2006).

Naturally derived coagulants include both the plant- and animal-based resources. Some of the more commonly used animal-based coagulants include isinglass from the shredded fish bladders (Biggs 2007) and chitosan from the shells of crustaceans (Bratby 2006). However, the available sources of plant-based coagulants are much higher than animal-based coagulants, thus suggesting that plant-based coagulants could be potential alternatives to chemical coagulants and have since gained gradual importance over the years.

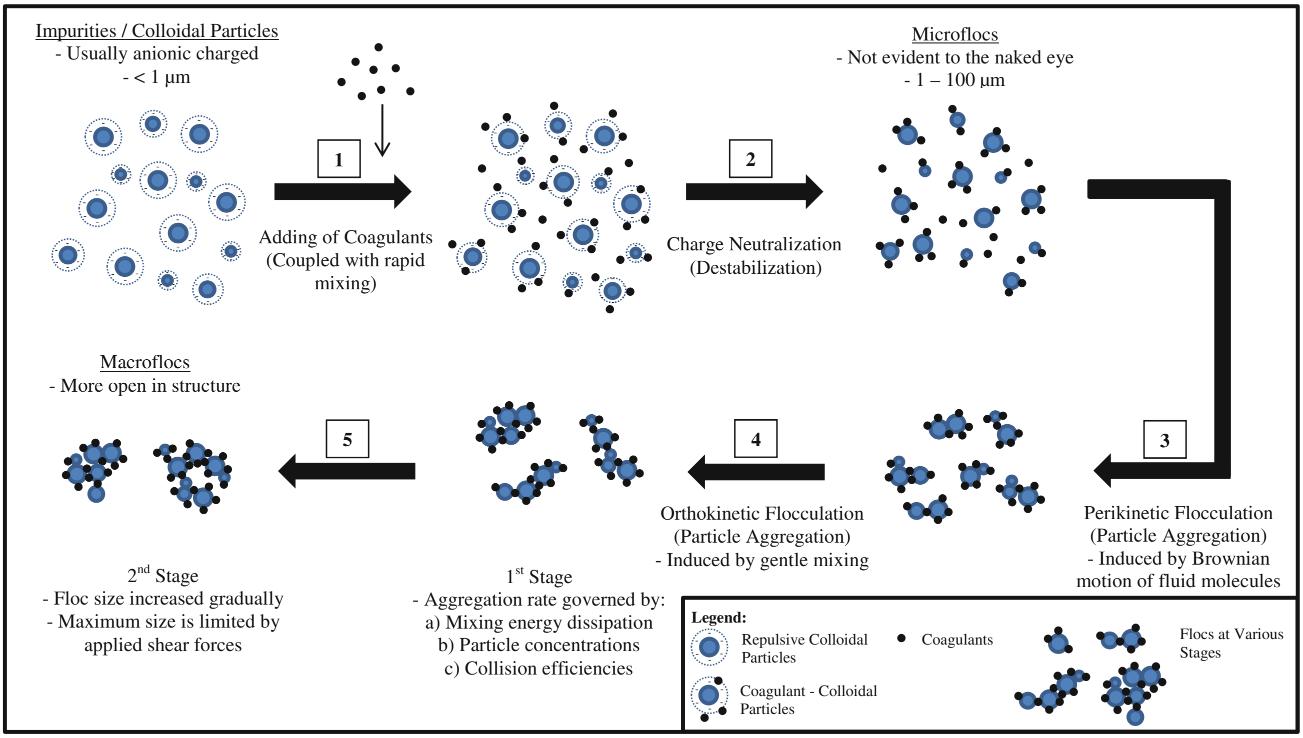


Fig. 1 Coagulation via charge neutralization and flocculation mechanism of the colloidal particles present in water

Recently, four main plant coagulants namely Nirmali seeds, *Moringa oleifera*, tannins and cactus that are more commonly known to the water and wastewater industries have been discussed in review papers (Yin 2010; Vijayaraghavan et al. 2011). Another review paper from Yongabi (2010) focused on the indigenous plant coagulants used by the rural Africans for water purification. An extensive review summarizing all the studied natural coagulants along with their additional benefits in the treated water is still lacking. Following this observation, the main motivation of this review paper is driven to compile the potential natural coagulants used and to identify the research gaps from the existing studies conducted. This work also aims to elucidate the coagulation mechanisms responsible for the observed turbidity removals when plant-based natural coagulants mainly vegetables and legumes were utilized in treating turbid waters.

Natural coagulants classified as common vegetables and legumes

The list of vegetables and legumes studied for their potential as natural coagulants have been summarized in Table 1 with brief accounts to their backgrounds, family classifications and common usages. Majority of the plant coagulants included in this review are legumes under the *Fabaceae* family which forms an economically important family of flowering plants.

The dimensions of the plant extracts will be handy for the packing process in terms of transportation and storage purposes when handled in bulk quantities. Also, larger-sized seeds are often favourable as more active coagulants are present relative to the increased seed size. The densities are important to gauge if the plant extracts are to be submerged entirely when added into the water samples for coagulation. The chemical compounds present in these plant coagulants such as protein and carbohydrates could have been responsible for their coagulation activities in water clarification. As such, it might be useful to link these values to the performance of the plant coagulants towards better understanding on their associated coagulation activities. Table 2 provides such information for comparisons among the various plant coagulants.

Table 3 summarizes the studies conducted in treating low to highly turbid water samples using the plant-based natural coagulants and the optimized parameters in achieving the highest turbidity removal.

Coccinia indica/Coccinia grandis

This herbaceous vine bearing small fruits (Table 1) is commonly referred to as ivy gourd and is a popular vegetable used for culinary purposes. The fruits which are rich in carbohydrates (Table 2) have been used in traditional medicine to cure diabetes, fever and leprosy (Lim 2012). Patale and Parikh (2010), Patale and Pandya (2012) used the mucilaginous extract obtained from this fruit for turbidity removal in synthetic water. Its mucilaginous extract was found to be effective in removing turbidity up to 94 % only in waters with turbidities of 100 NTU (Table 3). The natural anionic polysaccharide present in the mucilage is capable of promoting the formation of particle aggregates which could help in lowering turbidity.

However, the results reported were based on synthetic raw water. The usage of raw surface water would be ideal since it contains diverse types of nutrients, microorganisms as well as colloidal particles. The fruit extract was also reported to be rich in phenolic compounds and has been proven to exhibit antibacterial properties (Shaheen et al. 2009). One of the pathogenic bacteria *Salmonella parathyphi* could be present in the raw surface water due to the leaching of sewage effluents and can cause waterborne diseases. Thus, further study on this aspect is required when *Coccinia indica* is used as a natural coagulant.

Hibiscus esculentus/Abelmoschus esculentus

Hibiscus esculentus commonly referred to as lady's finger is widely consumed as a vegetable which is rich in protein and fibre. Its scientific name '*esculentus*' literally means edible and good to eat (Small 2011). The slimy mucilage obtained from the seed pods is used as effective natural coagulant in treating synthetic water and effluent (Anastasakis et al. 2009; Al—Samawi and Shokralla 1996; Agarwal et al. 2001, 2003). A wide range of initial turbidities has been tested up to 3000 NTU (Al—Samawi and Shokralla 1996) in synthetic water, which is crucial to provide an overview of the coagulation performance as high turbidities in surface water are possible especially after a heavy downpour or landslides. Initial turbidities up to 500 NTU are common in other studies conducted using natural coagulants. The performance of alum in turbid water of 100 NTU was enhanced with the addition of this seed extract, leading to higher turbidity removal up to 97.1 %. As a result, the usage of alum was minimized by 50 % (Al—Samawi and Shokralla 1996) and is economical especially for industrial applications.

The usage of seed pod extracts as a coagulant under optimized conditions removed more than 80 % of suspended solids from the effluents of sewage (Agarwal et al. 2001) and tannery (Agarwal et al. 2003). The X-ray diffraction analysis of the solid particles collected with and without the addition of the coagulant has shown differences in the values of 2θ angles and the d-spacing which corresponded to their interatomic spacing. It was then postulated

Table 1 Plant descriptions and background information for vegetables and legumes

No.	Scientific name	Common names	Family name	Brief plant descriptions	Usage	Country of origin	Ref.
1.	*Coccinia indica*	Ivy Gourd, Scarlet Gourd, Small Gourd, Kowai Fruit, Scarlet-Fruited Gourd	*Cucurbitaceae*	Robust and hardy aggressive herbaceous trailing vine with extensive root system. Highly adaptable to various habitat conditions	Medicinal purposes in India and Indonesia. Fruit, leaves and leafy shoots are consumed as vegetables	Central Africa, India and Asia	Lim (2012), Shaheen et al. (2009)
2.	*Hibiscus esculentus*	Okra, Lady's Finger, Gumbo, Gobo	*Malvaceae*	Annual herbaceous plant. Widely cultivated in the tropical and warmer countries. Often detested due to its sliminess	Consumed as a vegetable and used as a cooking ingredient. Gums as thickeners, emulsifying and foam stabilizing agents. Stems for paper and rope making	Old World tropics (West Africa)	Small (2011)
3.	*Luffa cylindrica*	Smooth Luffa, Egyptian Luffa, Vegetable Sponge, Sponge Guard	*Cucurbitaceae*	Vigorous climbing annual plant with fruits shaped like cucumbers but larger with a length of 30–60 cm. Green and sometimes mottled. Smooth with longitudinal lines	Fibrous interior as materials for filters, pillow stuffing and slippers. Immature fruits, leaves, flower buds and seeds are consumed	Old World tropics; probably Asia	Lim (2012), Small (2011)
4.	*Arachis hypogaea*	Peanut, Groundnut, Monkey Nut, Pinder, Goober	*Fabaceae*	Sparsely hairy annual herbaceous plant. Pod and seeds mature underground. Fruits begin as a fertilized flower above ground	Oilseed crop and food grain legume. Oil has widespread usages: pharmaceuticals, soaps, cold creams, plastics, dyes, massage oil and more	South America	Lim (2012), Boshou and Corley (2006), Fageria et al. (2010)
5.	*Cicer arietinum*	Dal Seeds, Chick Pea, Bengal Gram, Garbanzo Bean	*Fabaceae*	20–100 cm tall upright annual herb with pods of 14–35 mm long containing 1–2 (sometimes up to 4) seeds. Elongated or round seeds that are usually cream or yellow coloured	Food source as well as a traditional medicine. Good for soil conditionings	Mediterranean region	Lim (2012), Ahmad et al. (2005)
6.	*Dolichos biflorus*	Horsegram, Kulthi	*Fabaceae*	Climbing herb with stems that grows up to 60 cm in height. Pods contain 5–10 seeds that are usually pale to dark-reddish brown in colour	Used as cattle feed and can be consumed like any other beans. The stems and leaves served as fodder	Old World tropics	Brink (2006)
7.	*Glycine max*	Soybean, Soya Bean	*Fabaceae*	Bushy and can grow from 30 cm to 2 m tall. Pods contain 1–5 seeds (more commonly 2–3). Seeds are smooth, globule with varying colours depending on the type	Important food source of protein. Extracted oil has diverse applications: cooking oil, manufacture of soaps and other pharmaceutical products	Eastern Asia	Frederic Rosengarten (2004)
8.	*Guar gum*	Guar Bean, Cluster Bean, Guaran	*Fabaceae*	Robust plant growing up to 2 m but very susceptible to frost. 5–12 oval/cube shaped seeds in each pod with variable seed sizes	Seed gum as thickening and stabilizing agent in the food industry. Bean pods are consumed as vegetables	India	Peter Rory et al. (2001)
9.	*Lablab purpureus*	Hyacinth Bean, Bonavist Bean, Chink, Country Bean, Dolichos Bean	*Fabaceae*	Vigorous, herbaceous, short lived, twining vine that can grow up to 1 m in height. Deep-lavender purple or crimson coloured pods containing 3–5 seeds	Seeds consumed as green beans while leaves as spinach. Source of flour in pastry making. Herbage used as livestock feed	Old World tropics	Small (2009)
10.	*Phaseolus angularis*	Azuki Bean, Adsuki Bean, Red Bean	*Fabaceae*	Annual, bushy and erect herb which grows up to 90 cm high. Seeds are usually dark red; sometimes buff, straw-coloured, black or mottled	Dried seeds are consumed as a whole or processed into flour. Can also be popped like corn grains and leafy sprouts are eaten as vegetables	Exact origin is not known	Jansen (2006a)

Table 1 continued

No.	Scientific name	Common names	Family name	Brief plant descriptions	Usage	Country of origin	Ref.
11.	*Phaseolus mungo*	Urad Bean, Black Gram, Black Lentil, Black Matpe, Urd Bean	*Fabaceae*	Hairy, annual legume which can grow up to 100 cm high. Drought tolerant and can withstand dry areas. 4–10 black seeds per pod	Traditional medicine with various properties: narcotic roots suitable for inflammations, laxative seeds for constipation	India	Lim (2012)
12.	*Phaseolus vulgaris*	Common Bean	*Fabaceae*	Warm seasoned crop with an average of 4.4 seeds per pod based on various genotypes. Slender, twisted stems and branches	3rd most important legume worldwide. Consumed as a vegetable and a good source of protein	Central or South America	Fageria et al. (2010)
13.	*Pisum sativum*	Green Pea, Pea, Field Pea, Garden Pea, Stringless Snowpea	*Fabaceae*	Annual, bushy herb with stems up to 150 cm long. Ovate pods containing 3–11 seeds with varying shapes (spherical, angular to partially compressed)	Important source of protein and consumed fresh or processed. Can be used as stockfeed. Starch and fibre as additives in food processing industries	Southwestern Asia	Lim (2012)
14.	*Vigna unguiculata*	Cow Pea, Black Eyed-Pea, Southern Pea, Cowgram	*Fabaceae*	Annual herb with 12–30 cm long pods. 8–20 seeds per pod. Seeds could be speckled, mottled or botchy	Eaten as a vegetable. Exhibit medicinal properties: swellings and inflammations (leaves), insect stings (seeds), snake bites and chest pains (roots)	Southern Africa	Lim (2012)

that the primary bonding such as chelation between crystalline matter of the waste and the coagulant might have contributed to the agglomeration of the colloidal particles (Agarwal et al. 2001, 2003). The mucilage of *H. esculentus* consisted of both polysaccharide and protein fractions which have been found to exhibit strong interactions with one another (El-Mahdy and El-Sebaiy 1984). These fractions have molecular weights (MWs) of >100 kDa (El-Mahdy and El-Sebaiy 1984). On the other hand, commonly used anionic and non-ionic synthetic polymers have much higher MWs exceeding 1,000 kDa (Bolto and Gregory 2007). Thus, it is not surprising that the performance of natural polymers is somewhat less superior as opposed to those of chemical nature.

Turbidity removal in synthetic wastewater and biologically treated effluent was compared between *H. esculentus* and *Malva sylvestris*. The former exhibited a higher turbidity removal efficiency in the effluent sample at a much lower dosage (Anastasakis et al. 2009). However, the dissolved organic carbon content was increased in the treated effluent due to the addition of these plant coagulants (Table 3). The mucilage compositions of these plants might have influenced their resulting coagulation activity, and this warrants further study. The antimicrobial properties of this plant has shown potential against the inhibition to some of the tested bacteria such as *Escherichia coli* and *Staphylococcus aereus* which are commonly present in surface water (De Carvalho et al. 2011). Hence, it would be of great interest to study both its coagulation activity as well as the bacteria inhibition in the treatment of raw surface water for drinking purposes.

Luffa cylindrica/*Luffa aegyptiaca*

Due to its highly fibrous and sponge-like interiors, *Luffa cylindrica* is also known as sponge guard. This climbing plant is usually consumed as a vegetable when unripe as it lacks in the network of fibres (Small 2011). When the plant extract was added to raw surface water samples, turbidity reduction close to 85 % has been reported (Sowmeyan et al. 2011). As the exact part of the plant used was not specified, it can be postulated that either the seeds or the whole fruit exhibited such coagulative properties due to their similarities in terms of the high carbohydrate and protein contents (Table 2).

While most of the plant extracts undergo aqueous washing as part of the pretreatment process, Sowmeyan et al. (2011) introduced the formaldehyde or acid–alkaline wash. These solvents have been reported to be capable of removing organic matters and are commonly used in the ion exchange technology (Snoeyink and Scott Summers 1999). Hence, it can be assumed that some degree of organic matters from the plant extract has been removed

Table 2 Physicochemical properties for various vegetables and legumes (Seeds unless specified otherwise)

No.	Plant name	Physical Parameters						Proximate Compositions (% dry basis)					Ref.
		Length (mm)	Width (mm)	Breadth (mm)	Bulk density (kg/m^3)	True density (kg/m^3)	Porosity (%)	Moisture	Crude protein	Crude fibre	Carbohydrate	Fat/ lipid	
Vegetables													
1.	*Coccinia indica* Fruit[a]	30.0–70.0	10.0–35.0	–	–	–	–	94.0	20.0	26.7	51.7	1.7	Lim (2012)
2.	*Hibiscus esculentus*	5.2	4.8	4.1	592.0	1,107.0	46.3	6.4	19.1	26.3	–	8.2	Çalişir et al. (2005), Sahoo and Srivastava (2002)
3.	*Luffa cylindrica*[a]	10–120	–	–	–	–	–	4.4	26.8	6.8	32.0	26.8	Lim (2012), Dairo (2008)
	Luffa cylindrica Fruit[a]	100–500	50–100	–	–	–	–	93.9	19.6	–	72.0	3.3	Lim (2012)
Legumes													
4.	*Arachis hypogaea*[a]	11.2	7.6	6.9	–	1,010.0	–	5.8	40.7	3.9	1.9	49.5	Olajide and Igbeka (2003), Atasie et al. (2009)
5.	*Cicer arietinum* (split)[a]	6.3	5.3	2.9	713.0	1,202.0	40.7	9.8	18.2	16.4[b]	57.3	5.7	Lim (2012), Ghadge et al. (2008)
6.	*Dolichos biflorus*	3.0–8.0	3.0–5.0	–	–	–	–	8.0	22.1	5.6	37.2	–	Brink (2006), Ravindran and Bino Sundar (2009)
7.	*Glycine max*[a]	7.3	6.5	5.4	651.0	1,147.9	43.3	7.8	36.2	9.2[b]	30.0	19.7	Lim (2012), Tavakoli et al. (2009)
8.	Guar gum	–	–	–	–	–	–	8.0–14.0	3.0–6.0	1.0–4.0	73.0–86.7[c]	0.5–1.0[d]	Peter Rory et al. (2001)
9.	*Lablab purpureus*	–	–	–	–	–	–	6.4	26.9	–	67.2	1.9	Osman (2007)
	Lablab purpureus Peel[a]	40.0–50.0	–	–	–	–	–	9.8	16.4	37.3	–	–	Lim (2012), Shilpaa et al. (2012)
10.	*Phaseolus angularis*[a]	5.0–7.5	4.0–5.5	–	–	–	–	12.2	20.7	13.2[b]	65.5	0.5	Jansen (2006a)
11.	*Phaseolus mungo*[a]	5.0	–	–	–	–	–	9.1	23.4	17.0[b]	54.9	1.5	Jansen (2006b), Lim (2012)
12.	*Phaseolus vulgaris*[a]	5.0–15.0	–	–	–	–	–	10.6	22.4	24.0[b]	52.0	1.7	Wortmann (2006)
13.	*Pisum sativum*[a]	7.8	6.4	5.6	712.1	1,160.5	38.6	10.9	25.7	1.6	68.1	1.6	Lim (2012), Yalçin et al. (2007)

Table 2 continued

No.	Plant name	Physical Parameters						Proximate Compositions (% dry basis)					Ref.
		Length (mm)	Width (mm)	Breadth (mm)	Bulk density (kg/m^3)	True density (kg/m^3)	Porosity (%)	Moisture	Crude protein	Crude fibre	Carbohydrate	Fat/ lipid	
14.	*Vigna unguiculata*[a]	9.9	6.9	6.1	569.9	1,154.8	50.6	10.8	23.8	10.8[b]	60.9	1.3	Lim (2012), Yalçin (2007)

[a] Proximate compositions based on % dry basis except moisture were recalculated from the wet weight reported

[b] Dietary fibre

[c] Expressed as galactomannan

[d] Petroleum ether extractable

prior to the addition to the sample to reduce the leaching of organic matters. This is to be justified by measuring both the content of organic loadings in the sample before and after the addition of the plant extracts.

Antimicrobial properties in the aqueous extract of the powdered fruit as well as the seed extracts of *L. cylindrica* were observed against total and faecal coliform bacteria when added to raw surface water (Shaheed et al. 2009). The seed extracts which contained more antimicrobial constituents such as saponins and steroidal rings exhibited greater potency. The phytochemical profile as well as the antibacterial properties of the seeds and leaves has also been studied elsewhere (Oyetayo et al. 2007). However, the disinfection achieved for drinking water relying solely on this coagulant would not be sufficient (Shaheed et al. 2009). This plant extract has also been reported to remove heavy metals, fluoride and chlorides. Even so, it is more superior compared to the other studied coagulants in terms of removal of total dissolved solids (TDS) up to 60 % (Sowmeyan et al. 2011).

Arachis hypogaea

Arachis hypogaea or peanut is a good source of protein and is best known for its high oil content. The seeds are valuable in traditional medicine due to its anti-inflammatory properties, while the extracted oil is usually the basis of ointments (Lim 2012). The lipidic fraction in *A. hypogaea* contributes close to half of its dry weight (Table 2). Since lipid does not contribute to its coagulation activity, the relative concentration of the active agents is largely reduced, resulting in poorer turbidity removal in raw surface water (Table 3). Although the effect of delipidation is lacking, similar outcomes can be expected from the studies conducted on the seeds of *Moringa stenopetala* (Mataka et al. 2006). The delipidated cakes outperformed the crude seed extract in terms of heavy metal removal and the required coagulant dosage. Thus, it is strongly believed that the coagulation activity in *A. hypogaea* can be enhanced with the removal of this fraction.

Differences in the efficiency of turbidity removals between two studies were observed (Mbogo 2008; Subramanium et al. 2011). The characteristics of the raw surface water used in both the coagulation studies might have contributed to this contrast. The presence of high organic matters in water with greater surface charge has been found to dominate the coagulation process (Kim et al. 2001). Hence, it affected the neutralization of these colloidal particles and resulted in increased residual turbidity. The presence of multi-charged ions such as bivalent ions of magnesium and calcium could have aided the coagulation process. Studies have proven that the addition of these ions can help to reduce the residual turbidity (Tripathi et al.

Table 3 Research summaries of vegetables and legumes used as natural coagulants

No.	Coagulant		Type of sample	Initial conditions			Optimized parameters		Turbidity reduction (%)	Findings/claims	Ref.
	Name	Form used		Turbidity (NTU)	pH	Others	Dosage (mg/L)	pH			
1.	*Coccinia indica* fruit	B1, E, I, K, B2, F	SW (Kaolin)	100			0.4		94	Removal efficiency increased with increasing initial turbidity	Patale and Parikh (2010)
		B1, E, I, K, B2, F	SW (Kaolin)	100			0.4	7.5	94	Effective for turbidity removal at initial turbidities ≥100 NTU Efficiency decreased once the pH exceeded 7.5	Patale and Pandya (2012)
2.	*Hibiscus esculentus* seed pod tips, sap, plant stalk and roots	F, G, H, L	SW (Ball Clay Powder)	3,000	7.2–7.4		10		99.94	A more superior primary coagulant than: Alum at ≥900 NTU Nirmali seeds at ≥1,350 NTU Up to 90 % savings in alum when used as a coagulant aid	Al—Samawi and Shokralla (1996)
	Seed pods	L, K, B2, F	Sewage	225	7.6	SS: 165 mg/L	0.12	4.0		Removal of SS by 86.9 % Maximum solid removal from acidic to neutral condition	Agarwal et al. (2001)
		L, K, B2, F	TE	45.6	8.3	SS: 2,213 mg/L	0.04	9.2		Removal of SS by 98.3 % Removal of DS by 93.1 % Both acidic and basic conditions are suitable for solids removal	Agarwal et al. (2003)
		F, H, L	SWW (Kaolin)	63–73	5.9	DOC: 1.1 ppm	5		93–97.3	Mucilage which is also found in the *Opuntia* species contributed to its highly positive flocculating traits Increased DOC by 2.1 ppm	Anastasakis et al. (2009)
			BTE	55	6.9	DOC: 22.4 ppm	2.5–20		70–74	Increased DOC by 5.3 ppm	
3.	*Luffa cylindrica* (Not specified)	B3/B4, H	RSW	32	9.4	TDS: 606 mg/L	8,000 (B4)		~85	Most efficient for the removal of TDS	Sowmeyan et al. (2011)
4.	*Arachis hypogaea* seeds	F, H, L	RSW	482	7.6		1,000		96.74	Turbidity removal of up to 100 % when used as a coagulant aid (1,000 mg/L with 20 % alum)	Mbogo (2008)
		F, H, L	RSW (River)	~250					~24 (at 24th h)	Hardness removal of 25 % Lead removal of ~50 % Chromium removal of ~40 % Zinc removal of ~25 %	Subramanium et al. (2011)

Table 3 continued

No.	Coagulant		Type of sample	Initial conditions			Optimized parameters		Turbidity reduction (%)	Findings/claims	Ref.
	Name	Form used		Turbidity (NTU)	pH	Others	Dosage (mg/L)	pH			
5.	*Cicer arietinum* seeds	A, L	SW (Clay)	95			100		95.89	Total coliform counts reduced by 90.5 % Maximum turbidity removal in highly turbid water is comparable to alum	Asrafuzzaman et al. (2011)
6.	*Dolichos biflorus* seeds	F, H, I, J, M	SW (Clay)	100 mg/L			20	7.5	96	Maximum turbidity removal of 99.3 % at a dosage of 60 mg/L for initial turbidity of 1,000 mg/L	Thakre and Bhole (1985)
			SW (Clay)	100 mg/L			3 + 20 alum		99 (as CA which is added first)	Efficiency at different sequence of coagulant addition: 98 % with the simultaneous addition of both coagulants 95 % with the addition of alum followed by coagulant aid	
7.	*Glycine max* seeds	F, H, I, J, M	SW (Clay)	100			20	8.0	~77	Maximum turbidity removal of ~91 % at a dosage of 60 mg/L for initial turbidity of 1600 NTU	Bhole (1995)
			SW (Clay)	100			9 + 20 alum		~96.9 (as CA which is added first)	Efficiency at different sequence of coagulant addition: ~94 % with the addition of alum followed by coagulant aid ~92 % with the simultaneous addition of both coagulants	
		F, H, L	RSW	482	7.6		1,000		98.83	Turbidity removal of up to 100 % when used as a coagulant aid (800 mg/L with 20 % alum) More efficient than *Arachis hypogaea*	Mbogo (2008)
8.	Guar gum	L	RSW (Well)	49			50		96	Faecal coliforms removal of ~80 %. Very clear supernatant formed even at maximum coagulant dosage	Pritchard et al. (2009)
9.	*Lablab purpureus* seeds	F, H, I, J, M	SW (Clay)	100			20	7.0	~80	Maximum turbidity removal of ~81.3 % at a dosage of 60 mg/L for initial turbidity of 1,600 NTU	Bhole (1995)

Table 3 continued

No.	Coagulant		Type of sample	Initial conditions			Optimized parameters		Turbidity reduction (%)	Findings/claims	Ref.
	Name	Form used		Turbidity (NTU)	pH	Others	Dosage (mg/L)	pH			
			SW (Clay)	100			9 + 20 alum		~95.6 (as CA which is added first)	Efficiency at different sequence of coagulant addition: ~92 % with the addition of alum followed by coagulant aid ~91.5 % with the simultaneous addition of both coagulants	
		F, H, L	SW (Clay)	80			400		68.75 (at the 60th minute)	Further turbidity reduction when the finest grain size of coagulant is used in the medium to high turbidity water No bacterial growth even after a 24-h lag period → complete destruction in bacteria cells	Unnisa et al. (2010)
		F, C, H, L	SW (Clay)	100			100		88.9	Total coliform counts reduced by 89.5 %	Asrafuzzaman et al. (2011)
	Peels	F, H	SW	500			20	9.5	99.14	Less effective compared to *Opuntia ficus indica*	Shilpaa et al. (2012)
			RSW (Lake)	83			20		77.10	Bacterial reduction by 11.5 %	
10.	*Phaseolus angularis* seeds	F, H, D, L	SW (Kaolin)	250–300			10 µL/mL (in plastic cuvette)		~45–50	Salt extracted coagulant displayed a higher coagulation activity More or less similar coagulation activity up to 27 times dilution	Gunaratna et al. (2007)
11.	*Phaseolus mungo* seeds	F, H, L	RSW	482	7.6		800		100	100 % turbidity removal when used as a coagulant aid (600 mg/L with 20 % alum) Outperformed alum as primary coagulant	Mbogo (2008)
		F, H, L	RSW (River)	~250					~46 (at 24th h)	Lead removal of ~40 % Copper removal of ~30 % Chromium removal of ~25 %	Subramanium et al. (2011)
12.	*Phaseolus vulgaris* seeds	H, L	SW (Kaolin)	17.5			5	10	>75	High coagulation activity at pH 10 and above No coagulation activity at pH 7 Suitable for applications when initial turbidities ≤17.5 NTU	Šćiban et al. (2005)

Table 3 continued

No.	Coagulant		Type of sample	Initial conditions			Optimized parameters		Turbidity reduction (%)	Findings/claims	Ref.
	Name	Form used		Turbidity (NTU)	pH	Others	Dosage (mg/L)	pH			
		F, H, L, N F, H, L	SW (Kaolin)	35			1.0 mL/L 1.0 mL/L		72.3 30	Partially purified coagulant extract has: 22 times higher the coagulation activity 16 times lower organic load in treated water	Antov et al. (2010)
		H, L	SW (Kaolin)	35	9.0		3.5–4.5 (Samples 1,2,4) 1.5 (Sample 3)		~45 ~33	4 different strains of beans were used → Levač Bean—Sample 1, Sremac Bean—Sample 2, Zlatko Bean—Sample 3, Balkan Bean—Sample 4	Šćiban et al. (2010)
		H, L	DS (EJW)		4.2	COD: 66,850 mg O_2/L	5 mL/L	8.5		COD removal by 68.8 % With increasing coagulant dosage: COD removal increased at lower pH COD removal decreased at higher pH	Prodanović et al. (2011)
13.	*Pisum sativum* seeds	F, H, L	RSW	482	7.6		1,000		99.07	Turbidity removal of up to 100 % when used as a coagulant aid (800 mg/L with 20 % alum)	Mbogo (2008)
14.	*Vigna unguiculata* seeds	F, H, I, J, M	SW (Clay)	100			20	7.5	~83	Maximum turbidity removal of 82.1 % at a dosage of 60 mg/L for initial turbidity of 1,600 NTU	Bhole (1995)
			SW (Clay)	100			9 + 20 alum		~96.9 (as CA which is added first)	Efficiency at different sequence of coagulant addition: ~94 % with the addition of alum followed by coagulant aid ~92.5 % with the simultaneous addition of both coagulants	
		H, L	SW (Kaolin)	450–500	7.3 ± 1	Absorbance:1.3–1.6	12.5 µg/mL		~80	Wider pH range for significant coagulation activity in purified coagulant → pH 5.5–8.5 Optimum dosage is lowered 5–10 times in the case of purified coagulant	Marobhe et al. (2007)
		F, H, L	RSW	482	7.6		800		99.24	Turbidity removal of up to 100 % when used as a coagulant aid (800 mg/L with 20 % alum)	Mbogo (2008)

Table 3 continued

No.	Coagulant		Type of sample	Initial conditions			Optimized parameters		Turbidity reduction (%)	Findings/claims	Ref.
	Name	Form used		Turbidity (NTU)	pH	Others	Dosage (mg/L)	pH			
		F, H, L	RSW (River)	~250					~60 (at 24th h)	Chromium removal of ~40 % Lead removal of ~38 % Zinc removal of ~26 %	Subramanium et al. (2011)

A—Direct powder form, B—washed with: (1. Water, 2. Acetone, 3. Formaldehyde, 4. Acid-Alkaline), C—deshelled, D—oil removal with ethanol, E—cut, F—dried, G—removal of fibrous materials, H—pulverized, I—soaked in water, J—blended in water, K—precipitated with alcohol, L—aqueous extraction, M—added with HCl and N—partial purification

SW synthetic water, *TE* tannery effluent, *SWW* synthetic wastewater, *BTE* biologically treated effluent, *RSW* raw surface water, *CA* coagulant aid, *DS* distillery spent, *EJW* extraction juice wastewater, *SS* suspended solids, *DS* dissolved solids, *DOC* dissolved organic carbon, *TDS* total dissolved solids and *COD* chemical oxygen demand

1976; Okuda et al. 2001), while analogous effects have been observed when higher alkalinity and harder water samples were used (Muyibi and Evison 1996). These factors could justify the improved performance (Mbogo 2008) as the nature of the sample used also influences the end result of the coagulation treatment.

Not limited to water clarifications, this natural coagulant has been found to be capable of removing heavy metals. Apart from the listed metals, cadmium and copper were also removed despite much lower removal performances (Subramanium et al. 2011). These findings demonstrated its flexibility in treating turbid as well as waters contaminated with metals. The significantly high content of stearic and palmitic acid in *A. hypogaea* (Atasie et al. 2009) revealed the possibility of similar antibacterial properties to that in the seed pods of *H. esculentus*. This is so as both the plant extracts contained these saturated fatty acids which have been found to be responsible for the bacteria inhibition properties (De Carvalho et al. 2011). Hence, it can be postulated that some degree of *E. coli* removal is expected when this seed extract is tested on surface water samples although detailed studies have yet to be conducted.

Cicer arietinum

Apart from its traditional medicinal properties in the treatment of skin diseases and constipation (Lim 2012), *Cicer arietinum* or chick pea has been widely consumed as a food source which has high contents of carbohydrate and protein. Of late, this plant extract has also been found to exhibit coagulation activity in the treatment of synthetic water (Asrafuzzaman et al. 2011). However, like most of the natural coagulants, the effectiveness of *C. arietinum* in water clarification declined as the initial turbidity decreased. Maximum turbidity reductions of above 95 % for initial turbidity of 95 NTU was comparable to that in alum (Asrafuzzaman et al. 2011), and this is advantageous when the usage of alum is not favourable.

The chemical compositions in *C. arietinum* was found to be largely carbohydrate followed by crude protein (Table 2) which are the two most attributed constituents responsible for the coagulation of colloidal particles. Reduction in the total coliform has also been observed (Table 3) when the plant coagulant was added, indicating the possibilities of its antimicrobial activities. The presence of antifungal peptides with novel N-terminal sequences, designated as cicerin and arietin (Lim 2012), might have contributed to its potency against some of the bacteria. Thus, it would be beneficial to study its applications in the raw surface water to unleash its full potential as a natural coagulant.

Dolichos biflorus

Also known as horsegram, this climbing plant is usually made into silage. With its high nutritional content, especially protein and carbohydrates, the *Dolichos biflorus* seeds are also eaten as beans (Table 1). Besides its conventional usages, the seeds have been added as natural coagulants to treat synthetic water with initial turbidities ranging from 25 to 1,600 mg/L. Turbidity removals above 95 % in synthetic water under optimized conditions have been reported when used alone and in conjunctive usage with alum (Thakre and Bhole 1985), indicating its potential as an effective primary and coagulant aid. The plant extract worked best at pH values near neutral of 7.5 which is ideal considering the normal pH of surface water to be within 7–8 (Bhole 1995).

The order at which the coagulants were added has altered the treatment results (Table 3). If alum was first added, the alkalinity and pH levels in the sample would be reduced due to its working properties. When the extract of *D. biflorus* seeds was added next, its performance in promoting particle agglomerations declined as it has been reported to work well at pH near neutral. Thus, the resultant turbidity removal was lowered compared to the addition of the seed extracts followed by alum (Thakre and Bhole 1985). Despite its much lower protein content (Table 2), its overall performance as a natural coagulant is generally more efficient compared to *Vigna unguiculata,* the seeds of *Lablab purpureus* and *Glycine max* in the order of decreasing turbidity removals. The synergistic effects among the chemical constituents as well as the characteristics of the active coagulants present in each of the individual seeds might have contributed to this observation.

Glycine max

Widely known as soybean, the *G. max* plant is the most important source of vegetable oil, accounting for more than 50 % of the world's oilseeds (Frederic Rosengarten 2004). Its genus name '*Glycine*' has been derived with reference to the Greek word '*glykys*' which means sweet, thus indicating the sweetness of fruits from some of the species (Small 2009). Like most legumes, the seed extracts were reported to exhibit water clarification properties when tested in synthetic water (Bhole 1995) and highly turbid raw surface water (Mbogo 2008) as the primary coagulant and coagulant aid to alum. This plant coagulant is more superior in the treatment of surface water beyond 450 NTU (Mbogo 2008), suggesting its improved performance as the initial turbidity increases. Turbidity removals of >96 % (Table 3) have been observed when the plant extract was used as a coagulant aid to alum. This indicated an overall improvement compared to the sole addition of alum in water treatment.

The soybeans contained relatively large fraction of lipid and is the second highest legume trailing behind *A. hypogaea* (Table 2). This fraction does not contribute to coagulation activities, and delipidation of the seeds will be useful if enhancement in its turbidity removal is required as the relative concentration of the active agents has been increased. In addition to turbidity removal, delipidated or deoiled soybeans have also been recently studied as low-cost bio-adsorbents in the treatment of various dye-contaminated water (Mittal et al. 2008, 2009a, b, 2010a, b, c; Gupta et al. 2009). Palmitic and stearic acids which contributed to the bactericidal activities in *H. esculentus* (De Carvalho et al. 2011) are also present in *G. max* (Lim 2012). Hence, this plant extract could also exhibit potency against some of the bacteria present in raw surface water.

Guar gum from *Cyamopsis tetragonoloba*

The endosperm splits of seeds from the sun-loving plant of *Cyamopsis tetragonoloba* or commonly known as cluster beans produce the guar gum flour (Peter Rory et al. 2001). This food gum is 'Generally Recognized as Safe' by the US Drug and Food Administration (Melnick et al. 1983), and little evidence was linked to its adverse human health impacts (Peter Rory et al. 2001). Thus, this plant extract is safe for human consumption when used as a natural coagulant. The treatment of water samples with initial turbidities below 50 NTU obtained from five shallow wells has been studied with the addition of guar gum (Pritchard et al. 2009). Surprisingly, a 100 % turbidity removal was reported for an initial turbidity of 1 NTU despite the removal trend to be somewhat affected by the degree of initial turbidity. Even so, its overall performance when treating higher-initial turbidity waters up to 49 NTU fared better than *Jatropha curcas* but less superior compared to *M. oleifera* (Pritchard et al. 2009).

Guar gum is extremely low in protein content if compared to other legume sources which acted as natural coagulants (Table 2), suggesting that protein might not be the dominating active agents responsible for its coagulative activities. In fact, guar gum which is a neutral polysaccharide (Bratby 2006) is highly rich in carbohydrate in the form of galactomannan. Guar gum also has high molecular weights of 50,000–8,000,000 (Mirhosseini and Amid 2012). This large polymeric structure is also more polydisperse compared to another natural coagulant, the locust bean gum (Lawrence 2003). As such, it can be postulated that guar gum is more superior in removing turbidity considering the much easier dispersion and at least 2.5 times greater in terms of molecular weight (Mirhosseini and Amid 2012).

The faecal coliform contents in the treated water also showed significant reductions. However, this does not comply with the requirements of achieving zero colony-forming units (cfu)/100 mL as stipulated by WHO, and subsequent disinfections are necessary to ensure adequate water sanitation (Pritchard et al. 2009). Bacteria might have been removed along with the colloidal particles, and the antimicrobial properties of guar gum demands additional research to identify the possible chemical constituents leading to its bacteria inhibitions.

Lablab purpureus

The *L. purpureus* plant often bears lavender-purplish flowers, thus contributing to its genus name '*purpureus*' (Small 2009). This robust growing creeper commonly known as hyacinth bean (Table 1) has been studied as a primary coagulant as well as an aid to alum (Bhole 1995). Turbidity removal in synthetic water was increased by 15 % when added as a coagulant aid (Table 3). At 1,600 NTU, further turbidity removal of up to 82 % was reported (Bhole 1995). Its effectiveness in treating turbid water was greatly enhanced when used as a coagulant aid. This could provide an alternative means in bid to reduce the dependency on chemical coagulants. In general, the peels of *L. purpureus* (Shilpaa et al. 2012) can be postulated to be more efficient as a relatively small dosage is capable of achieving higher turbidity removal.

The turbidity removals in two relatively similar initial conditions of the studied synthetic water (Unnisa et al. 2010; Asrafuzzaman et al. 2011) varied by at least 20 % (Table 3). Both the sedimentation times were approximately 60 min, but the coagulant dosage required was four times larger when shelled seeds were used (Unnisa et al. 2010). This clearly implied that the shell removal of *L. purpureus* seeds is beneficial as it increases the relative concentrations of the active coagulant compounds. Thus, higher turbidity removal can be achieved while reducing the coagulant dosage. This is also in close agreement with the usage of deshelled *M. oleifera* seeds as natural coagulants which have shown similar trends (Ndabigengesere and Subba Narasiah 1998a, b). The chloroform and n-hexane extracts of *L. purpureus* have been found to be significantly potent against the range of bacteria tested (Lim 2012). As aqueous extractions of natural coagulants are commonly adopted, its antimicrobial activity in this solvent would be useful particularly for the treatment of drinking water.

Phaseolus angularis

Phaseolus angularis, widely known as red bean, is high in proteins and carbohydrate (Table 2). The active protein coagulant was found to be of cationic nature with similar properties to that in the seeds of *M. oleifera* (Gunaratna et al. 2007). However, its turbidity removal in synthetic water was outclassed by the latter, which could be due to its 1.7 times lower protein content (Compaoré et al. 2011) despite having larger protein molecular weight. Even though the carbohydrate content is about 7.3 times greater (Compaoré et al. 2011), this fraction which is also responsible for the particle coagulation does not seem to have much influence on the corresponding activity. The limited presence of the branched polysaccharides such as amylopectin which leads to effective coagulation in the roots extract of *Maerua subcordata* (Mavura et al. 2008) could also be accounted for the decreased coagulation activity. Along with its reduced protein content, it is not surprising that *M. oleifera* is more efficient as the active agents have been identified to be that of protein nature (Ndabigengesere et al. 1995).

The resulting coagulation activity was sustained regardless of the number of dilution factors (Gunaratna et al. 2007). This is economically beneficial as both the diluted and concentrated protein extracts can achieve relatively similar coagulation activities. In fact, the diluted protein extracts showed improved coagulation activity that was lower compared to *M. oleifera*. Gunaratna et al. (2007) also discovered that the protein coagulant was stable at elevated temperatures beyond 85 °C with 30 % enhanced activity, but its turbidity removal remained fairly low, which is undesirable for the treatment of drinking water.

The tannin constituent from the seeds of *P. angularis* exhibited antibacterial activities against *E. coli* and other bacteria which are the common causes of waterborne illnesses in surface water (Amarowicz et al. 2008). Polyphenols present in this coloured seeds were proposed as the possible cause leading to bacterial inhibitions (Hori et al. 2006). However, the susceptibility towards polyphenols is dependent on the characteristics of the individual microorganisms. Its conjunctive usage as a natural coagulant for both turbidity and microorganism removal warrants further research to gauge its full potential if it is to be adopted in the existing water treatment technology.

Phaseolus mungo/*Vigna mungo*

In India, the *Phaseolus mungo* plant or black gram is a pulse crop that is largely grown for the production of dhall (Lim 2012). This annual plant is also grown for forage and has widespread applications as a traditional medicine. Recently, its potential as a natural coagulant has been studied in treating highly turbid raw surface water (Mbogo 2008; Subramanium et al. 2011). The seed extracts were capable of removing 100 % turbidity at a reduced dosage by 20 % to outperform alum as a primary coagulant

(Mbogo 2008). Also, *P. mungo* was found to be the most efficient natural coagulant compared to *V. unguiculata*, *Pisum sativum*, *G. max* and *A. hypogaea* in the decreasing order of efficiency in water clarification. This could be due to its higher protein content (Table 2) with <2 % of lipid fraction. The cationic nature of its polyelectrolytes as well as protein (Subramanium et al. 2011) is essential for the neutralization of the colloidal particles which promoted the formation of flocs.

The seed extracts of *P. mungo* have additional advantage when used in water treatment. Heavy metals such as lead, copper and chromium were removed by more than 25 % along with smaller removal fractions of hardness, zinc as well as cadmium (Subramanium et al. 2011). This is beneficial when such metals are present in raw surface water and must be removed to meet the drinking water quality. However, it lacks superiority in comparison with the extracts of *M. oleifera* (Subramanium et al. 2011). Other heavy metal removal techniques such as the use of carbon nanotubes as adsorbents could be employed to enhance their removal in contaminated water (Gupta et al. 2011a, b).

Phaseolus vulgaris

Also known as common bean, this plant is the most widely distributed and consumed seed legume in the species of *Phaseolus* globally (Fageria et al. 2010). Its expanded usages is not only limited to providing traditional remedies against diabetes mellitus (Lim 2012), but also as a potential natural coagulant in removing turbidity. However, its turbidity removals in treating lowly turbid synthetic water of <40 NTU (Antov et al. 2010; Šćiban et al. 2010) have been found to be below 50 % (Table 3). This is so as the findings by Šćiban et al. (2005) showed that the seed extracts of *Phaseolus vulgaris* performed well at pH values beyond 10 and has good coagulation activities at turbidities below 18 NTU. Since the water conditions were not adjusted to the optimized treatment conditions, the performance of the seed extracts have yet to reach the pinnacle indicating the importance of pH control. This extract is useful when treating such lowly turbid water as turbidity removals >75 % (Šćiban et al. 2005) have been reported.

Partial purification of these protein coagulants have also been studied and compared against the coagulation activity of the crude proteins (Antov et al. 2010). This additional pretreatment step is highly important to lower the introduction of organic loadings (Ghebremichael 2007) which is a common issue when using natural coagulants and to reduce the dosage of coagulant in achieving similar coagulation activities. The molecular weight of the trimer coagulant protein subunit is approximately 50 kDa which is larger than that in *M. oleifera*. As the seeds of *P. vulgaris* contained a much smaller fraction of lipid, this eliminates the need for delipidation and is highly beneficial in terms of economic and environmental concerns (Antov et al. 2010).

Two commercial treatment processes, centrifugation and coagulation using plant coagulants, were compared for various distillery wastewaters. The usage of the seed extracts in the extraction juice wastewater sample yielded COD removals up to 69 % under optimized parameters as opposed to centrifugation (Prodanović et al. 2011).

Pisum sativum

Pisum sativum or pea is rich in carbohydrate and protein, which makes it an important food source that is consumed fresh or as raw material in the canning industry. This bushy plant also served as a traditional relief to various skin problems, and with good gel strength, its starch is used in the production of packaging materials (Lim 2012). Positive results have been reported when the aqueous extract of the seeds were used to clarify highly turbid surface water (Mbogo 2008). At a similar coagulant dosage of 1,000 mg/L, improved performance compared to *G. max* and *A. hypogaea*, both having higher protein contents by at least 1.4 times (Table 2), has been observed (Mbogo 2008). It can then be anticipated that their resulting coagulation activity would also be greater considering the active agents contributing to coagulation are often proteins.

However, the mixture of chemical constituents in the extracts could have synergistic effects towards coagulation and the carbohydrate fraction consisting of starch could also be one of the coagulation agents. Starch, a non-ionic polymer, could have been responsible for its relatively high turbidity removal apart from the protein coagulant. This explains its improved coagulation efficiency whereby the larger fraction of carbohydrate has dominating effect towards particle aggregations. The coagulation activity in the plant extracts such as those from the roots of *M. subcordata* (Mavura et al. 2008) and seeds of *Zea mays* (Raghuwanshi et al. 2002) has also been attributed to the presence of starch.

Good bacteria inhibition against 56 isolates belonging to 11 different species of Gram-negative bacilli has been reported (Saeed and Tariq 2005). The growth of pathogenic bacteria commonly present in the raw surface water such as *E. coli* and *Salmonella spp*. could be effectively retarded as the seeds are rich in phenolic antioxidant (Saeed and Tariq 2005). As the juice of fresh seeds (Saeed and Tariq 2005) was used, differences in the resulting bacteria inhibition may exist when the aqueous extract (Mbogo 2008) is tested on contaminated surface water due to the variations in the working form. However, this study has provided a platform towards the potential for microorganism removal when used as a natural coagulant.

Vigna unguiculata

Vigna unguiculata or cow pea has long been recognized as a major food staple, especially in most of the African countries (Lim 2012). Besides its traditional medicinal applications (Table 2), the seed extracts were reported to exhibit coagulation activities when tested on turbid synthetic water (Bhole 1995; Marobhe et al. 2007) and raw surface water (Mbogo 2008; Subramanium et al. 2011). Generally, turbidity removals of >80 % can be attained with the usage of these seed extracts regardless as a primary or as a coagulant aid (Table 3), but the result presented in one of the studies fell below this minimum percentage (Subramanium et al. 2011). It is likely due to the absence of dosage optimization as the turbidity removal is not maximized. Turbidity removals were enhanced at higher initial turbidities as observed for most of the natural coagulants.

The coagulation activities in the crude and purified protein coagulant from the seeds of *V. unguiculata* have been studied under various conditions (Marobhe et al. 2007). The purified cationic proteins through ion-exchange chromatography have similarities with the protein coagulant in *M. oleifera*. Two or more coagulant proteins were reported to be present in this extract (Marobhe et al. 2007). Purification of the protein coagulant enabled significant dosage reductions in achieving comparable results to that in the crude protein with the potential of removing the associated organic load introduced in the treated water. Both the crude and purified extracts of *V. unguiculata* could retain at least 85 % of the coagulation activities when heated at 40 °C but became highly sensitive after prolonged heating at much higher temperatures of above 80 °C (Marobhe et al. 2007). Given the lower temperature of the surface water, coagulation activities would not be affected with the usage of this plant coagulant.

The extracts were also capable of removing <25 % of hardness, cadmium and copper apart from its water clarification activities (Subramanium et al. 2011). Although its bacteria inhibitions in surface water are still lacking, studies have concluded the presence of various proteins with antifungal and antiviral potency in the seeds of *V. unguiculata* (Lim 2012), which may lead to some form of water disinfection properties.

Coagulation/destabilization mechanisms

Coagulation or destabilization of the suspended colloidal particles can be achieved via four mechanisms, namely the double-layer compression, charge neutralization, bridging as well as sweep coagulation (Crittenden et al. 2005). The coagulation mechanisms work based on different principles and one or more mechanisms could be employed for more effective particle destabilizations.

- Double-layer compression

This coagulation mechanism relied on the addition of an ‘indifferent’ electrolyte in large quantities. The high ionic solution introduced in an existing system would alter the overall ionic concentration. Thus, the double layer surrounding the colloidal particle would be compressed to a certain extent where the repulsive energy barrier will also be lowered. This phenomenon will encourage the binding of two molecules and subsequently aid in the formation of flocs. However, the effectiveness of this coagulation mechanism is questionable and is usually not preferred. The presence of bivalent ions such as Ca^{2+} and Mg^{2+} in water has been attributed to induce some form of coagulation activities via the double-layer compression mechanism (Duan et al. 2009).

- Charge neutralization

This coagulation mechanism involves the adsorption of an oppositely charged coagulant on the colloidal surface. The colloidal particles are usually negatively charged under normal surface water conditions. Hence, positively charged coagulants will be attracted towards the colloids, subsequently resulting in surface charge neutralizations. The quantification of zeta potential is highly beneficial in predicting the optimum coagulant dosage required to achieve effective coagulation. A near-zero net charge would provide a clear indication of the ideal coagulant dosage sufficient for complete charge neutralizations. As such, the effectiveness of this mechanism is strongly dependent on the coagulant dosage introduced; particle restabilization could easily occur once the optimum dosage is exceeded. The cationic protein fraction from natural coagulants such as the seeds of *M. oleifera* (Ndabigengesere et al. 1995) and *J. curcas* (Abidin et al. 2011) has previously been reported to work under this coagulation mechanism.

- Adsorption and bridging

The bridging of particles occurs with the introduction of long-chain polymers or polyelectrolytes as these coagulants are capable of extending into the solution to capture and bind multiple colloids together. The bridging efficiency is further improved when coagulants with larger molecular weights are used due to the extended polymeric chains. Natural polymers such as polysaccharides and proteins could also induce coagulation via bridging. The mucilage of cactus *Opuntia ficus indica* which is an anionic polysaccharide has been proven to result in particle removals via this mechanism (Miller et al. 2008).

- Sweep coagulation

The formation of coagulant precipitates via the addition of an unusually large coagulant dosage is the backbone leading to sweep coagulation. These particles could act as nucleation sites to facilitate the precipitation formation (Sincero and Sincero 2002). As the precipitate forms, colloidal particles would be enmeshed in the growing precipitate and thus could be successfully removed from the colloidal suspension. Sweep coagulation could result in improved coagulation for greater removal performance in comparison with charge neutralization (Duan and Gregory 2003). As a far greater coagulant dosage than that for the latter is required, the larger amount of sludge generated at the end of the coagulation process may not be favourable. Unlike chemical coagulants such as alum, plant-based natural coagulants may not employ sweep coagulation as the turbidity reductions observed generally declined at elevated coagulant dosages beyond the optimum values (Agarwal et al. 2001; Al—Samawi and Shokralla 1996).

Table 4 provides a concise summary of the key features and characteristics of each coagulation mechanisms. The floc characteristics have been compiled from the results obtained using chemical coagulants specifically alum. However, the same can be anticipated for plant-based natural coagulants due to the relevance of the appropriate coagulation mechanisms. Based on the nature and type of natural coagulants, the main coagulation mechanisms responsible for the observed turbidity removals (Table 3) could most likely be charge neutralizations and bridging.

It is also important to note that the performances of these two coagulation mechanisms are strongly affected by pH as well as the coagulant dosage. Hence, subsequent optimization studies would be worthwhile to maximize the full potential of plant-based natural coagulants in achieving the highest turbidity reductions. The significance of these critical parameters will be discussed further in the following subsections.

Influence of pH on turbidity reductions

The isoelectronic point (pI) and pH of a solution are two important factors affecting the overall particle charge. A significant number of studies conducted on plant-based natural coagulants have prepared synthetic turbid water via the addition of clay or kaolin particles (Table 3). As the pI of kaolin has been reported to be 2.8 (Ndabigengesere and Subba Narasiah 1996), the kaolin particles were negatively charged at the tested water samples. The polysaccharide fraction specifically mucilaginous extracts from *C. indica* and *H. esculentus* has been recognized as potential natural coagulants based on the relatively high turbidity removals reported. The predominant coagulation mechanism leading to particle destabilizations could be postulated to be adsorption and bridging owing to the anionic nature of both the mucilage.

The pH optimization studies have been conducted on selected vegetables: *C. indica* (Patale and Pandya 2012), *H. esculentus* (Agarwal et al. 2001, 2003) and legumes (Thakre and Bhole 1985; Bhole 1995; Shilpaa et al. 2012; Šćiban et al. 2005; Prodanović et al. 2011). The manipulation of pH can significantly enhance the efficiency of coagulation process as pH alters the electrochemical nature of both the solvent and ionic polymers used (Somasundaran et al. 2005). However, the effects of pH fluctuations are more prominent in polymers with a high degree of ionic charges. Although non-ionic natural polymer such as guar gum is less sensitive to pH variations, a much greater coagulant dosage can be expected in contrast to the usage of ionic natural polymers (Biggs 2007). This is evident from the optimized coagulant dosage of guar gum where the anionic mucilage of *C. indica* and *H. esculentus* required at least five times lower dosages despite being used at higher initial turbidities (Table 3). Yet, neutral polymers would be ideal for the treatment of water with higher hardness or salinity levels (Biggs 2007).

The two active coagulation agents identified from vegetables and legumes as natural coagulants explicitly polysaccharides and proteins are made up of long polymeric chains. Based on the similarity to the structure of organic polyelectrolytes, the usage of natural polymers can also be correlated to two main controlling factors resulting in coagulation: the conformation and charge properties of polymers (Yu and Somasundaran 1996). Both the mucilage of *H. esculentus* and *Opuntia ficus indica* contained galacturonic acid which has been found to be an active coagulation agent (El-Mahdy and El-Sebaiy 1984; Miller et al. 2008). The charge density of galacturonic acid which has a weak carboxylic acid group attached to the polymeric chain is pH dependent (Bolto and Gregory 2007). The carboxylate group of the anionic mucilage is prone to ionization with increments in pH. As a result, the uncoiling and stretching of the polymeric chain is enhanced due to the amplified intramolecular electrostatic repulsions induced by the ionization of the acidic group (Yu and Somasundaran 1996).

The optimized pH for enhanced coagulation activities using the mucilage of *H. esculentus* lied between the mildly acidic and basic regions (Table 3). Generally, a lower solid removal efficiency has been observed at a neutral pH of 7 (Agarwal et al. 2003). At this pH value, the relative mucilage viscosity was reported to be at its peak (Woolfe et al. 1977). Hence, it would not be surprising that reduced coagulation activities were observed due to the increased difficulty in mucilage dispersion. A random coil configuration is most often adopted when polymers are used in water treatment (Bolto and Gregory 2007). At a higher pH of 9, the anionic mucilage of *H. esculentus* would uncoil further to attain a flat, dangling form which is beneficial for coagulation enhancements. This is so as a larger quantity of unoccupied spaces on the polymeric chain is introduced for improved bridging of the colloidal particles. Studies have also concluded that the relative mucilage viscosity can be lowered with increasing concentrations of bivalent ions (Woolfe et al. 1977). Owing to the possible variations in particle compositions between the sewage wastewater and tannery effluent, the optimized pH was obtained in two different regions.

Apart from polysaccharide, proteins which are amphoteric molecules are also greatly affected by pH. Most of the legumes used as natural coagulants worked best at optimized pH of 7–8.5 (Table 3). The two predominant amino acids present in the seed legumes were reported to be glutamic acid and aspartic acid (Dhawan et al. 1991; Kovalenko et al. 2006; Audu and Aremu 2011; Padhye and Salunkhe 1979; Leterme et al. 1990; Hussain and Basahy 1998). These acidic amino acid side chains have low pI values of below 3.3 (Properties of Amino Acids 2010), contributing to the overall anionic protein surface charge at the optimized pH values. As such, charge neutralization via the smaller fractions of basic side chains present in the seed legumes, namely lysine, arginine and histidine with pI values larger than 7.6, is most likely the secondary coagulation mechanism.

Generally, residual turbidities of the treated water increased once the optimum pH has been exceeded (Thakre and Bhole 1985; Bhole 1995; Shilpaa et al. 2012). As the positivity of the individual surface charges declined, the extent of charge neutralization would be reduced. However, the peels of *L. purpureus* and seed extract of *P. vulgaris* were more superior at increased basicity with pH values >9 (Shilpaa et al. 2012; Šćiban et al. 2005). Polymer-induced coagulation is a complex process which is often affected by the roles of various parameters such as the characteristics of solvents and polymers used as well as their interactions on the process behaviours (Somasundaran et al. 2005). The coiling ratio of a polymer with the changing pH of the solution could be obtained via fluorescence spectroscopy to gauge the most probable polymeric structure (Yu and Somasundaran 1996). This aspect of studies would be useful to verify the postulated coagulation mechanisms at any given pH regions.

Influence of dosage on turbidity reductions

The dosing of coagulants is critical as the process requires care and proper control to achieve the desired quality of treated water. Often at times, poor coagulant dosage results in unsatisfactory treated water quality, owing to the lack of process optimizations. Hence, it is vital to determine the optimum range of coagulant dosage required to achieve maximum turbidity removals at a minimized treatment cost. Among the four coagulation mechanisms, only charge neutralization and bridging are negatively affected by coagulant dosage due to the stoichiometric relationship exhibited (Table 4).

Three possible dosing cases can be obtained, namely under dosing, optimum dosing and over dosing. Figure 2 depicts the effects of coagulant dosage with regard to the efficiency of particle agglomerations via charge neutralization and bridging. Particle restabilization leading to a decline in turbidity removal efficiency can be expected once the optimum coagulant dosage is exceeded. Additional adsorption of the cationic coagulants on the 'readily neutral' colloidal particles would lead to charge reversals and subsequently result in the repulsion of particles hindering particle agglomerations. Overcrowding of coagulants in a solution would also limit the amount of adsorption sites available for particle bridging as complete surface coverage on the coagulant has occurred (Bolto and Gregory 2007). Thus, the addition of more coagulants would not improve the coagulation process further. In contrast, a much-lower-than-required coagulant dosage would lead to incomplete and ineffective coagulation as a majority of the colloidal particles are left in suspension (Fig. 2).

The performance of natural coagulants in turbidity removal over a selected range of coagulant dosage has previously been studied: *C. indica* (Patale and Parikh 2010), *H. esculentus* (Al—Samawi and Shokralla 1996; Agarwal et al. 2001, 2003; Anastasakis et al. 2009) and various legumes (Mbogo 2008; Asrafuzzaman et al. 2011; Thakre and Bhole 1985; Pritchard et al. 2009; Shilpaa et al. 2012; Šćiban et al. 2005, 2010; Antov et al. 2010; Prodanović et al. 2011; Marobhe et al. 2007). Even when natural coagulants were used as coagulant aids, dosage optimization is equally important to enhance the overall water treatment efficiencies (Bhole 1995).

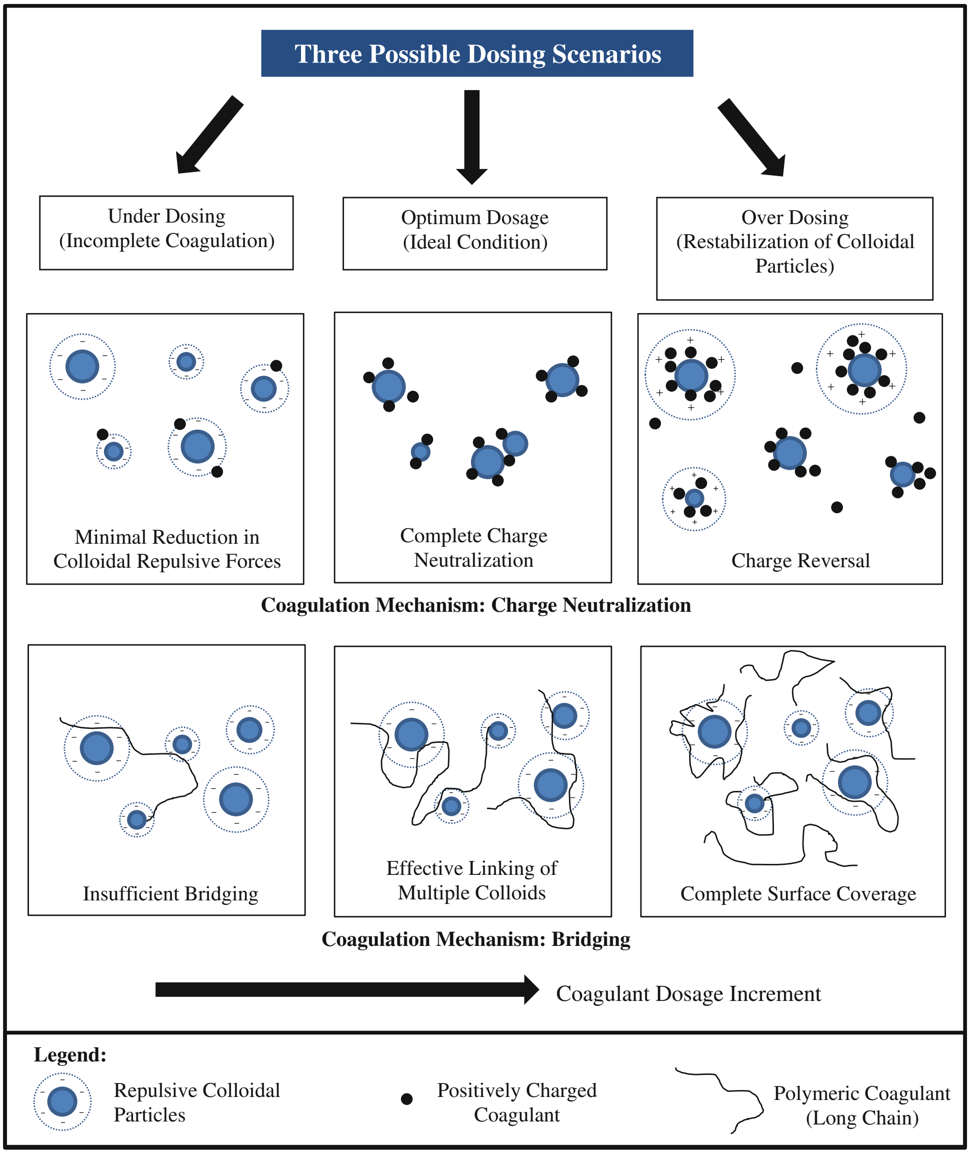
The concentration of colloidal particles held in suspension is directly related to the initial turbidity of a given water sample as a high initial turbidity translates to a large amount of suspended particles. As such, the coagulant dosage required for complete destabilization via charge neutralization and bridging would somewhat increase proportionally with the concentration of the particles (Table 4). Gradual increments in the coagulant dosage approaching the optimum range have positive effects on enhancing turbidity removals (Marobhe et al. 2007; Patale and Parikh 2010; Thakre and Bhole 1985). However, a surge in residual turbidity has been observed at coagulant dosages beyond the bench point as represented by the optimum dosage,

Table 4 Comparisons of various coagulation mechanisms

	Double-layer compression	Adsorption and charge neutralization	Adsorption and bridging	Sweep coagulation (precipitation)	Ref.
Key features of coagulation mechanisms					
Brief description of mechanism	Compression in double layer with the addition of ionic solutions	Attraction between oppositely charged ions leading to overall surface charge neutralization	Linking of multiple colloids via the addition of polymeric coagulants	Entrapment of colloids within the growing precipitates	
Additional attribute	Charge density of ionic solution affects the degree of double-layer compression introduced	At the optimum coagulant dosage, Zeta Potential ~ 0	Works well with polyelectrolyte of high molecular weight due to increased length of the polymeric chain	Sludge volume increased proportionately to the dosage of coagulants added	Ndabigengesere et al. (1995), Biggs (2007), Duan and Gregory (2003)
Colloidal concentration and dosage	No relationship between optimum dosage and colloid concentration	Stoichiometry relationship between particle concentration and optimum coagulant dosage	Stoichiometry relationship between particle concentration and optimum coagulant dosage	No relationship between optimum dosage and colloid concentration	Bratby (2006)
Coagulant dosage required	Occurs at high concentrations of ionic solution	Usually occurs at low coagulant dosage sufficient for charge neutralization	Occurs at low coagulant dosage (0.0625 mg/L polyacrylamide)	Occurs at elevated coagulant dosage ($\sim$18.8 times of charge neutralization)	Li et al. (2006)
Floc characteristics					
Floc size	Small in size	Smallest: Diameter $\sim$[a] $G^{-0.6017}$	Largest: Diameter $\sim$[a] $G^{-0.3674}$	Moderate in size: Diameter $\sim$[a] $G^{-0.5618}$	Li et al. (2006)
Floc strength	Weak owing to the electrostatic forces	Weakest: Up to 0.43 N/m^2	Strongest: Up to 0.58 N/m^2	Stronger than charge neutralization flocs: Up to 0.56 N/m^2	Li et al. (2006)
Fractal dimension	Larger than bridging	Larger than bridging: 2.55–2.76 (2nd most compact)	Smallest due to the loosely attached flocs: 2.40–2.63	Largest as flocs are the most compact: 2.58–2.91	Li et al. (2006)
Floc structure	Compact, denser	Less dense, more open structure	Three-dimensional open structure	Compact, denser	Bratby (2006)
Floc shape	Close to spherical	Relatively spherical	Long, threadlike	Close to spherical	Miller et al. (2008)
Breakage reversibility		Usually reversible as van der Waals forces can happen anytime leading to higher floc recovery	Partial recovery due to the shearing of long-chain polymers into various sections which may be irreversible	Irreversible and reduced recovery as bonding sites becomes unavailable	McCurdy et al. (2004)

[a] G represents the average velocity gradient

Fig. 2 Effects of coagulant dosage on charge neutralization and bridging



which could be justified by the restabilization of particles due to the overcrowding effect of coagulants (Fig. 2). The optimized natural coagulant dosage corresponded to the highest turbidity removal achieved (Table 3) would serve as a reference when working with such coagulants for water clarifications. Although the surface water characteristics and other process variables may vary, both the polysaccharide and protein fractions as active coagulation agents from vegetables and legumes require precise dosing to attain the desired turbidity removal efficiencies.

Current research gaps and limitations

The growing importance on minimizing environmental implications has led to changes in both the energy and waste management systems (Nouri et al. 2012), making way for the emergence of cleaner productions while highlighting pollution prevention (Wu et al. 2010). Plant-based natural coagulants could be a worthwhile alternative in addressing the environmental and ecological concerns raised over the usage of chemical coagulants in water clarification. Despite interesting research findings including the recent work carried out on mustard (Bodlund et al. 2013), which suggest to their immense potential as future coagulants, certain aspects of the coagulation studies are still in infantry. The respective coagulation mechanisms for most of the natural coagulants have yet to be understood clearly unlike the widely developed chemical coagulants. Often at times, the active agents responsible for coagulations are made up of different chemical constituents. It is likely that synergistic effects between the polypeptides and polysaccharides in natural coagulants have resulted in the observed coagulation activities. However, this would warrant further purification and isolation of the respective active proteins or polysaccharides in grasping the underlying coagulation principles at work. Difficulties in postulating the coagulation mechanism were further heightened with the lack of information, especially in terms of zeta potential measurements. The availability of such data could provide direct indications to the most probable coagulation mechanisms owing to their distinctive features as discussed in Coagulation/destabilization mechanisms section.

While various studies have highlighted the superiority of natural coagulants in inducing particle agglomerations (Table 3), the resultant flocs formed have yet to be analysed and characterized based on floc size, shape and extent of compactness. Such information on floc characteristics could reveal important evidence to support the postulated coagulation mechanisms. It is also vital to determine the floc strength in order to gauge the durability of the floc in resisting breakage. The flocs formed would be subjected to shearing forces in the subsequent water treatment processes if natural coagulants are to be used commercially. Little attention has also been given to the optimization of pH as one of the major parameters affecting the overall efficiencies in turbidity reduction. Natural coagulants would perform differently when tested over a wide range of pH, and this governs the need to select the ideal pH at which maximum turbidity reduction can be attained. As pH plays a vital role in determining the solubility and dominant surface charges of the active coagulation agents, a balance between the extent of pH adjustments needed and the subsequent turbidity removal efficiencies should be obtained.

Conclusion

Some of the plant-based natural coagulants presented in this review paper have shown remarkable results and could be adopted either as primary coagulants or coagulant aids. Particle destabilizations resulting from the use of vegetables and legumes were induced via charge neutralization and bridging. Although numerous studies have utilized such extracts for turbidity removals, commercialization and industrial applications are still lacking. They have the potential to be explored further as alternatives in clarifying turbid water. In-depth studies on the characterizations of the active coagulant compounds would be beneficial to gain the necessary knowledge in understanding their respective coagulation activities. The efficiency of natural coagulants could be heightened with the optimization on both the pH and coagulant dosage used. Gradual introduction to the existing water treatment technology is possible once the bottlenecks of commercialization and limitations of natural coagulants have been resolved.

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